

# Critical dynamics of singlet and triplet excitations in strongly frustrated spin systems

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New data for the two-dimensional  $J_1 - J_2$  - model shows that the critical points for singlet and triplet excitations near  $J_2/J_1 \approx 0.38$  are very close together, or coincident. We propose that this is a more general result: in strongly frustrated spin systems the critical dynamics of singlet and triplet excitations are closely related. An effective field theory for such a critical point, or points, is developed, and its implications discussed. The analysis suggests the existence of gapless or almost gapless singlet excitations in the magnetically disordered phase.

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Quantum phase transitions in quantum antiferromagnets have attracted a great deal of attention during the last decade. This field contains numerous previously unknown phenomena. Investigation of quantum phase transitions in quantum antiferromagnets is very important for quantum magnetism, but is of even wider importance because it allows one to construct and to analyze effective nonperturbative field theories that can be used in other parts of physics. In this paper we concentrate on rotationally invariant (2+1)D systems. Transitions described by the nonlinear  $O(N)$  field theory are very well understood, see, e.g., Refs. [1–3]. The most widely known example is the  $O(3)$  theory that describes a real vector field  $\mathbf{v}$  with Lagrangian density

$$\mathcal{L}_v = \frac{1}{2} [(\partial_t \mathbf{v})^2 - c_v^2 (\nabla \mathbf{v})^2 - m_v^2 \mathbf{v}^2] - \frac{u_v}{4} (\mathbf{v}^2)^2. \quad (1)$$

The effective mass vanishes at some critical point,  $m_v^2 = a_v(g - g_{cv})$ , where  $g$  is some generalized “coupling constant”, and  $a_v$  is some coefficient. It has been argued by Haldane [4] and Chakravarty, Halperin and Nelson [5] that this theory describes a quantum phase transition between the Néel state ( $g < g_{cv}$ ) and the magnetically disordered state ( $g > g_{cv}$ ) in a two-dimensional quantum antiferromagnet. Perhaps the simplest concrete example of such a system is the Heisenberg antiferromagnet on a double layer square lattice. In this case the “coupling constant” is equal to the ratio of interlayer and inlayer antiferromagnetic interactions,  $g = J_\perp/J$ ,  $g_{cv} \approx 2.525$ , see Ref. [6]. For  $g > g_{cv}$  the excitation spectrum of the system (1) is gapped, and for  $g < g_{cv}$  the spectrum is gapless because of Goldstone spin waves.

The  $O(1)$  field theory describes a real scalar field  $s$  with Lagrangian

$$\mathcal{L}_s = \frac{1}{2} [(\partial_t s)^2 - c_s^2 (\nabla s)^2 - m_s^2 s^2] - \frac{u_s}{4} s^4, \quad (2)$$

where  $m_s^2 = a_s(g_{cs} - g)$ . The field expectation value  $\langle s \rangle$  is zero at  $g < g_{cs}$  and nonzero at  $g > g_{cs}$ . The spectrum is gapped in both cases. A simple example of such a system is a Heisenberg antiferromagnet on a “planar pyrochlore” lattice where the entire low energy dynamics is driven by spin singlet excitations [7].

The situation in strongly frustrated spin systems is much more complicated because the dynamics of triplet and singlet excitations are not independent. The 2D  $J_1 - J_2$  model is an ideal testing ground for understanding of such dynamics. The Hamiltonian of the model reads:

$$H = J_1 \sum_{nn} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{nnn} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (3)$$

where  $J_1$  is the nearest-neighbor, and  $J_2$  is the frustrating next-nearest-neighbor Heisenberg exchange on a square lattice. Both couplings are antiferromagnetic, i.e.  $J_{1,2} > 0$  and the spins  $S_i = 1/2$ . At small  $J_2$  the system exhibits Néel ordering and at small  $J_1$  it exhibits collinear ordering. There is also a general consensus on the disappearance of the magnetic ordering at  $0.38 < J_2/J_1 < 0.6$ , see, e.g., Ref. [8]. The structure of the magnetically disordered state is of great interest. An important insight into the disordered regime was achieved by field-theory methods [9,8] and dimer series expansions [10,11,8]. The above works have shown that the ground state in this regime is dominated by short-range singlet (dimer) formation in a columnar pattern. More recent studies based on the Green function Monte Carlo method favored either a plaquette state [12] or a dimer state with plaquette modulation [13]. And finally the same group that claimed stability of the plaquette state [12], now claims stability of the translationally invariant RVB state [14]. The above series expansions [10,11,8], as well as Monte Carlo numerical calculations [12–14] in various forms, have difficulties in distinguishing between states with very close energies, as occurs in the  $J_1 - J_2$  model. In such approaches one starts from some trial wave function (dimer, plaquette, RVB,...) and such an ansatz is always very restrictive: it is impossible to get a translationally invariant state using the dimer or plaquette expansions, and on the other hand it is impossible to get a spontaneous violation of the translational symmetry starting from the trial RVB state that is explicitly translationally invariant.

A novel approach was implicitly formulated in our work [15], see also Ref. [16]. The idea of the approach is to use numerical calculations only in the well established Néel phase at  $J_2/J_1 < 0.38$ . This calculation allow us to find

all the relevant quasiparticles in the system, and hence to determine the effective field theory describing the dynamics of the system. Since the transition is of the second order the same field theory describes the magnetically disordered phase and we have no problem to describe this phase. In the present work we explicitly formulate the approach, present new, much more accurate, numerical data, derive the effective Lagrangian of the model, and finally discuss implications of this effective Lagrangian.

For the  $J_1 - J_2$  model we define the generalized coupling constant as  $g = J_2/J_1$ . It is well established that at  $g < g_{cv}$  there is Néel ordering in the system. There are 3 independent estimates for  $g_{cv}$ . The first comes from dimer series expansions,  $g_{cv} \approx 0.38$ , see Ref. [8]. However this estimate is problematic since it already assumes spin dimer order at  $g > g_{cv}$ . The second estimate,  $g_{cv} = 0.4 \pm 0.02$ , comes from Ising series expansions for the staggered magnetization in the Néel phase [11]. The third estimate  $g_{cv} = 0.38 \pm 0.03$  comes from the Ising series expansions for spin-spin correlators in the Néel phase [15]. Using the second and the third estimates, and averaging them as independent ones we find

$$g_{cv} = 0.39 \pm 0.016 \rightarrow 0.39 \pm 0.02. \quad (4)$$

The Néel ordering corresponds to the condensation of a vector field, therefore there is no doubt that some effective Lagrangian determines the dynamics of the field both at  $g < g_{cv}$  and  $g > g_{cv}$ .

However the vector excitation is not the only relevant degree of freedom in the system. There are also the low energy singlet excitations. We do not have a reliable numerical technique for direct calculation of singlet excitations, but we do have a well developed series expansion technique for calculation of static susceptibilities. A static susceptibility is proportional to the corresponding Green's function at zero frequency  $\chi_{\mathbf{q}} \propto G_{\mathbf{q}}(\omega = 0) \sim Z_{\mathbf{q}}/\omega_{\mathbf{q}}^2$ , where  $\omega_{\mathbf{q}}$  is the quasiparticle energy, and  $Z_{\mathbf{q}}$  is the quasiparticle residue. So at the critical point  $1/\chi$  must vanish approximately as  $(g - g_c)^\gamma$ , with  $\gamma = \nu(2 - \eta)$ , where  $\nu$  is the critical index for the spectral gap,  $\Delta \propto (g - g_c)^\nu$ , and  $\nu\eta$  is the critical index for the quasiparticle residue,  $Z \propto (g - g_c)^{\nu\eta}$ . For the simple O(1) Lagrangian (2) the critical index is  $\gamma \approx 1.2$  (see, e.g., Ref. [2]).

In this paper we use Ising series expansion [11] techniques to compute the susceptibility of this system. The linked-cluster expansion method has been reviewed in a recent article [17], and will not be repeated here.

In Ref. [15] we have calculated the susceptibility of the Néel state with respect to the external field.

$$F_d = f \sum_{i,j} (-1)^i \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j}, \quad (5)$$

where the coefficient  $f$  gives the strength of the field. The susceptibility is defined in the usual way

$$\chi = \frac{1}{2} \frac{\partial^2 E}{\partial^2 f} \bigg|_{f=0}, \quad (6)$$

where  $E$  is the ground state energy. This susceptibility probes spin singlet excitations that correspond to a spin columnar dimerization. The susceptibility is diverging at  $g = J_2/J_1 \sim 0.4$  and this is a clear indication of the critical singlet excitation.

Unfortunately convergence of the series expansion for  $\chi$  is somewhat erratic and therefore the error bars in the numerical data [15] are rather large. In the present work we calculate susceptibilities  $\chi_z$  and  $\chi_\perp$  with respect to the fields

$$\begin{aligned} F_{dz} &= f \sum_{i,j} (-1)^i S_{i,j}^{(z)} S_{i+1,j}^{(z)}, \\ F_{d\perp} &= f \sum_{i,j} (-1)^i \left( S_{i,j}^{(x)} S_{i+1,j}^{(x)} + S_{i,j}^{(y)} S_{i+1,j}^{(y)} \right). \end{aligned} \quad (7)$$

Here  $z$  is direction of the staggered magnetization in the Néel state. Both  $\chi_z$  and  $\chi_\perp$  probe the same excitations as  $\chi$ . Using the Ising expansion about the Néel order [11], we have computed the series for  $\chi_z$  and  $\chi_\perp$  in powers of exchange anisotropy to order 7 for given values of  $J_2/J_1$ . The calculations involve a list of 28474 distinct connected clusters up to 9 sites. We do not display the series here but can provide them on request. Fortunately convergence of the series for  $\chi_\perp$  and  $\chi_z$  is much better than that for  $\chi$ . Let us note that the field  $F_d$  can be represented as  $F_d = F_{dz} + F_{d\perp}$ . However the susceptibility  $\chi$  cannot be represented as a simple superposition of  $\chi_z$  and  $\chi_\perp$ ,  $\chi \neq \chi_z + \chi_\perp$ . Therefore a relatively poor convergence of the series for  $\chi$  does not contradict the better convergence for  $\chi_z$  and  $\chi_\perp$ . The results for  $1/\chi_z$  and  $1/\chi_\perp$  are presented in Fig.1 and Fig.2. The susceptibilities are diverging at the critical point  $g_{cs}$ . From Fig.1 we conclude that  $g_{cs} = 0.38 \pm 0.04$ , and from Fig.2 we conclude that  $g_{cs} = 0.41 \pm 0.03$ . Considering these estimates as independent ones and averaging them we find

$$g_{cs} = 0.40 \pm 0.024 \rightarrow 0.40 \pm 0.02. \quad (8)$$

We have also calculated the susceptibility  $\chi_p$  with respect to the field

$$F_p = \sum_{i,j} (-1)^{i+j} \left( S_{i,j}^{(x)} S_{i+1,j+1}^{(x)} + S_{i,j}^{(y)} S_{i+1,j+1}^{(y)} \right), \quad (9)$$

that probes excitations of plaquette symmetry, but this is not coupled to the spin-dimer excitations. The Ising series for  $\chi_p$  has been computed up to order 7. The calculations involve a list of connected clusters different from that for  $\chi_z$  and  $\chi_\perp$  because the symmetry of the field is different. The number of cluster in this calculation is 28450, also up to 9 sites. The results of the Ising series expansion calculations for  $1/\chi_p$  are shown in Fig.3. According to the data the plaquette excitation is not critical and hence is not relevant.

The susceptibilities (7) probe the real scalar field  $s$  that corresponds to the spin columnar dimerization. The ground state expectation value of the field is zero at  $g < g_{cs}$  (no spontaneous dimerization), but the susceptibilities are diverging at  $g_{cs}$ . Therefore one can say the effective Lagrangian for the field  $s$  is given by Eq. (2). This is a possible scenario, but does not explain why the critical points for the scalar field (8) and for the vector field (4) coincide or at least are very close: in this scenario this is just a pure coincidence. To be absolutely correct we have to say that there is another scalar field  $s'$  that corresponds to the replacement  $i \rightarrow j, j \rightarrow i$  in Eqs. (7). So the symmetry of the transition is  $O(1) \times O(1)$ . However this additional  $s'$ -field is completely identical to  $s$ , and therefore it does not require a special consideration.

The fields  $\mathbf{v}$ ,  $s$  and  $s'$  are not independent: they certainly interact with each other. Therefore there is an alternative scenario that naturally explains the coincidence of the quantum critical points. Consider the effective Lagrangian of an interacting massive vector field and two massless scalar fields.

$$\begin{aligned} \mathcal{L} = & \frac{1}{2} [(\partial_t \mathbf{v})^2 - c_v^2 (\nabla \mathbf{v})^2 - m_v^2 \mathbf{v}^2] - \frac{u_v}{4} (\mathbf{v}^2)^2 \\ & + \frac{1}{2} [(\partial_t s)^2 - c_s^2 (\nabla s)^2 + (\partial_t s')^2 - c_s^2 (\nabla s')^2] \\ & - \frac{u_s}{4} (s^2 + s'^2)^2 - \frac{u'_s}{4} s^2 s'^2 - \frac{u_{sv}}{4} \mathbf{v}^2 (s^2 + s'^2), \end{aligned} \quad (10)$$

where  $m_v^2 = a_v(g - g_{cv})$ . This Lagrangian incorporates all the relevant degrees of freedom. At  $g < g_{cv}$  the vector field develops the ground state expectation value  $\langle \mathbf{v} \rangle = \sqrt{a_v(g_{cv} - g)/u_v}$ . and, hence, because of the interaction the massless scalar fields attain an effective mass  $m_{eff}^2 = a_v(g_{cv} - g)u_{sv}/(2u_v)$ . Thus in this scenario the scalar susceptibilities are diverging exactly at the same point where the Néel order disappears,  $\chi \propto 1/(g_{cv} - g)$ . In the magnetically disordered phase at  $g > g_{cv}$  both the scalar fields remain massless. This scenario corresponds to a translationally invariant spin liquid state. One can also introduce in Eq. (10) a small negative bare mass for the scalar fields,  $m_s^2 = -|m_s^2|$ . However due to Eqs. (4) and (8) this mass must be very small because it splits the critical points. In this case there is a region between the critical points where the spin-dimer and the Néel order coexist [15,16].

We mention, in this connection, that in one-dimensional systems the Lieb-Schultz-Mattis (LSM) theorem guarantees that a uniform phase in a quantum spin system is gapless and a gapped phase breaks translational symmetry [18]. Extension of the LSM theorem to two dimensions was proposed [19] but not proven (see also Ref. [20]). We note that the effective Lagrangian (10) is in perfect agreement with the conjectured two-dimensional extension of the LSM theorem. If the bare singlet mass  $m_s^2$  is zero then the magnetically disordered state is uniform and there are two gapless excitations in the state. If

$m_s^2 = -|m_s^2| \neq 0$  then one of the scalar fields develops the ground state expectation value  $\langle s \rangle = \sqrt{|m_s^2|/u_s}$  (spontaneous dimerization) and another attains finite positive mass due to the interaction terms,  $m_{eff}^2 = \frac{u_s}{2u_s}|m_s^2|$ .

The effective Lagrangian (10) corresponds to the translationally invariant intermediate spin liquid state (SL) and in this part it is in agreement with the conclusion of the recent work [14]. However, as we have shown, this SL state must have gapless singlet excitations. Therefore: 1) the dimer-dimer correlation function must fall off as  $\propto 1/r$ , where  $r$  is the separation between dimers, 2) for a finite cluster of  $N$  sites the static susceptibility with respect to the dimerization field (5) must scale as

$$\chi = \frac{1}{2f^2} \frac{|\langle 0|F_d|s \rangle|^2}{E_0 - E_s} \propto \frac{1}{f^2} \frac{(f\sqrt{N})^2}{(1/\sqrt{N})} \propto N^{3/2}. \quad (11)$$

Both these conclusions disagree with results of Ref. [14]. In our opinion this can be related to the variational nature of that calculation. We would like to stress that for calculation of the susceptibility the external field must be weak,  $f\sqrt{N} \ll E_s - E_0 \sim 1/\sqrt{N}$ . If this criterion is violated, the ground state energy shift is proportional to  $\propto f\sqrt{N}$  and the second derivative (6) vanishes.

It is more likely that the bare singlet mass  $m_s^2 = -|m_s^2|$  is small, but non-zero. The dimer series expansions [8] clearly show two nonzero dimer order parameters in the SL phase. To avoid misunderstanding we stress that we do not claim that one can derive a translationally invariant ground state starting from the dimer basis. This is certainly impossible. However if the true state is a translationally invariant one then the dimer series must generate zero dimer order parameters as an indication that there is something wrong with the dimer state. This does not happen. In principle it is certainly possible that convergence of series is so peculiar that the standard Pade approximant method based on a limited number of terms does not give the right answer, but this is an unlikely event and there are no examples of such behavior. In the case of the dimer ground state the static susceptibility of a finite cluster with respect to the dimerization field (5) must scale as

$$\chi = \frac{1}{2f^2} \frac{|\langle 0|F_d|0 \rangle|^2}{\Delta E} \propto \frac{1}{f^2} \frac{(fN)^2}{\exp(-aN)} \propto N^2 e^{aN}. \quad (12)$$

For the calculation the external field must be very weak,  $fN \ll \Delta E \sim \exp(-aN)$ . If this criterion is violated, the ground state energy shift is proportional to  $\propto fN$  and the second derivative (6) vanishes.

In conclusion we have demonstrated that the critical dynamics of singlet and triplet excitations in the two-dimensional  $J_1 - J_2$ -model are closely related. The critical points for singlet and triplet excitations are very close or coincide. This is a new type of critical behavior and

we argue that this is a generic property of strongly frustrated spin systems. We have formulated the effective field theory describing the critical behavior. The analysis favors gapless or almost gapless singlet excitations in the magnetically disordered phase. The effective field theory deduced in our work supports the two-dimensional extension of the Lieb-Schultz-Mattis theorem conjectured some time ago.

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- [1] J. Zinn-Justin *Quantum field theory and critical phenomena* (Oxford University Press, Oxford, 1993).
  - [2] R. Guida and J. Zinn-Justin, J. Phys. A **31**, 8103 (1998).
  - [3] S. Sachdev, in *Quantum phase transitions* (Cambridge University Press, Cambridge, 1999).
  - [4] F. D. M. Haldane, Phys. Rev. Lett. **50**, 1153 (1983).
  - [5] S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. Lett. **60**, 1057 (1988); Phys. Rev. B **39**, 2344 (1989).
  - [6] K. Hida, J. Phys. Soc. Jpn. **59**, 2230 (1990); Zheng Weihong, Phys. Rev. B **55** 12267 (1997); P. V. Shevchenko, A. W. Sandvik, and O. P. Sushkov, Phys. Rev. B **61**, 3475 (2000).
  - [7] V. N. Kotov, M. E. Zhitomirsky, and O. P. Sushkov, Phys. Rev. B **63**, 064412 (2001).
  - [8] V. N. Kotov, J. Oitmaa, O. P. Sushkov, and Zheng Weihong, Phil. Mag. B **80**, 1483 (2000).
  - [9] N. Read and S. Sachdev, Phys. Rev. Lett. **66**, 1773 (1991); *ibid.* **62**, 1697 (1989).
  - [10] M.P. Gelfand, R.R.P. Singh, and D.A. Huse, Phys. Rev. B **40**, 10801 (1989); M.P. Gelfand, *ibid.* **42**, 8206 (1990).
  - [11] J. Oitmaa and Zheng Weihong, Phys. Rev. B, **54**, 3022 (1996); R.R.P. Singh, Zheng Weihong, C. J. Hamer, and J. Oitmaa, Phys. Rev. B **60**, 7278 (1999).
  - [12] L. Capriotti and S. Sorella, Phys. Rev. Lett. **84**, 3173(2000).
  - [13] M. S. L. du Croo de Jongh, J. M. J. van Leeuwen, and W. van Saarloos, Phys. Rev. B **62**, 14844 (2000).
  - [14] L. Capriotti, F. Becca, A. Parola, and S. Sorella, Phys. Rev. Lett. **87**, 097201 (2001).
  - [15] O. P. Sushkov, J. Oitmaa, and Zheng Weihong, Phys. Rev. B **63**, 104420 (2001).
  - [16] S. Sachdev and Kwon Park, cond-mat/0108214.
  - [17] M.P. Gelfand and R.R.P. Singh, Adv. Phys. **49**, 93(2000).
  - [18] E. Lieb, T. Schultz, and D. Mattis, Ann. Phys. (N.Y.) **16**, 407 (1961).
  - [19] I. Affleck, Phys. Rev. B **37**, 5186 (1988).
  - [20] G. Misguich and C. Lhuillier, cond-mat/0002170.

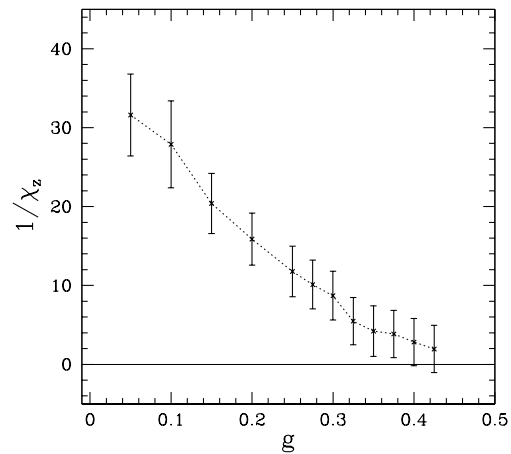


FIG. 1. The plot of  $1/\chi_z$ , where  $\chi_z$  is the dimer susceptibility calculated in the Néel state using Ising series expansion. The value of  $1/\chi_z$  vanishes at  $g_{cs} = 0.38 \pm 0.04$ .

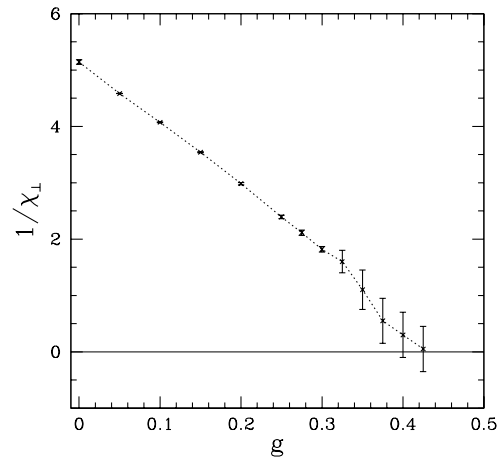


FIG. 2. The plot of  $1/\chi_{\perp}$ , where  $\chi_{\perp}$  is the dimer susceptibility calculated in the Néel state using Ising series expansion. The value of  $1/\chi_{\perp}$  vanishes at  $g_{cs} = 0.41 \pm 0.03$ .

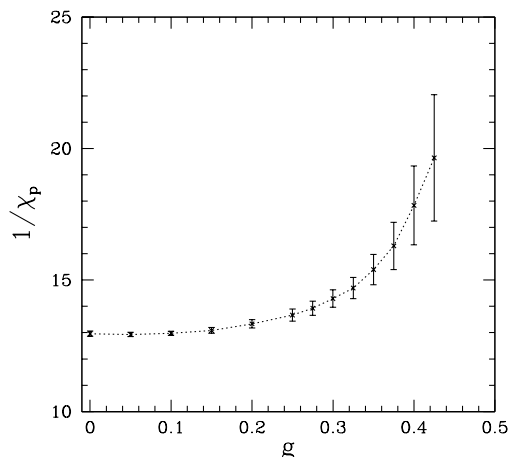


FIG. 3. The plot of  $1/\chi_p$ , where  $\chi_p$  is the plaquette susceptibility calculated in the Néel state using Ising series expansion. The value of  $1/\chi_p$  does not vanish, so excitations with plaquette symmetry are not critical.